REMARKS

Claims 16-41 are all the claims pending in the application. Claims 16, 17, 18, 19, 21, 36, and 37 have been amended for purposes of clarity or to correct typographical errors. In addition, claims 16 and 37 have been amended based on, for example, pages 38-39 of the specification. New claims 38-41 have been added to further define the dielectric barrier discharge.

In addition, the specification has been amended to change "CU" to "Cu".

Entry of the above amendments is respectfully requested.

Initially, Applicants thank the Examiner for indicating that claims 24-25 would be allowable if rewritten in independent form.

I. Response to Objection to Specification and Claims

On page 2 of the Office Action, the Examiner sets forth guidelines for the preferred layout for the specification of a utility application. Applicants thank the Examiner for providing the guidelines. However, 37 C.F.R. § 1.77(b) suggests, and does not require, the particular sections in the particular order. Accordingly, Applicants have not amended the specification and withdrawal of the objection is respectfully requested.

In addition, claims 16, 18, and 21 are objected to because in claim 16 line 16, the term "the metal surface" lacks antecedent basis, and in claims 18 and 21, the element should be "Cu". Claims 16 and 36 have been amended to provide proper antecedent basis. Claims 18 and 21 have been amended to change "CU" to "Cu".

In view of the above, withdrawal of the objections is respectfully requested.

II. Response to Rejection of Claims 16, 19, 30-31, and 34-37 under 35 U.S.C. § 112 second paragraph

Claims 16, 19, 30-31, and 34-37 are rejected under 35 U.S.C. §112, second paragraph, as being indefinite.

Applicants respectfully traverse the rejection.

The Examiner asserts that in claims 16, 19, 36 and 37, the mechanisms in the parenthetical elements are deemed vague and indefinite as to whether these are required by the applicant or not.

Without acquiescing the merits of the rejection, claims 16, 19, 36 and 37 have been amended to delete "(silent discharge or creeping discharge)" and new dependent claims 38-41 have been added.

Next, the Examiner asserts that in claim 16, lines 17-18, the term "taking out" is deemed vague and confusing as to where it is taking out from.

It is respectfully submitted that one of skill in the art would understand "taking out" as meaning "removing". However, for purposes of further clarity, claims 16 and 17 have been amended to change "taking out" to "removing".

The Examiner asserts that in claims 16, 19, 36 and 37, the term "high-field" is a relative term which renders the claim indefinite because the term "high" is not defined by the claim and the specification does not provide a standard for ascertaining the requisite degree.

It is respectfully submitted that "high-field intermittent discharge plasma" is a term of art that that would be understood by one of skill in the art. To support Applicants' position, three technical publications are submitted herewith.

The Examiner asserts that in claims 30 and 31, the terms "large quantity", "large ...

surface" are relative terms which renders the claim indefinite because the term "large" is not defined by the claim and the specification does not provide a standard for ascertaining the requisite degree.

Without acquiescing the merits of the rejection, the term "large" has been deleted from claims 30 and 31.

Finally, the Examiner asserts that in claims 34 and 35, the phrase "by an ozonizer that provided on a previous stage" is vague and confusing.

It is respectfully submitted that claims 34 and 35 are clear and that one of skill in the art would understand that an ozonizer is provided in the line that supplies oxygen from the oxygen cylinder. Specifically, on page 38 of the specification, it is disclosed that an ozonizer 710 is provided in the line 10 that supplies oxygen from the oxygen cylinder 8, and the gas that is once transformed to high-concentration ozonized oxygen gas 101 by the ozonizer is supplied to the photocatalyst material producing apparatus. Nonetheless, claims 34 and 35 have been amended for purposes of further clarity.

In view of the above, withdrawal of the rejection is respectfully requested.

III. Response to Rejection of Claims 16-18, 22, 28, 30, 32, 34 and 36 under 35 U.S.C. § 103(a)

Claims 16-18, 22, 28, 30, 32, 34, and 36 are rejected under 35 U.S.C. §103(a) as allegedly being unpatentable over Tabata et al. (US 2004/0223893).

Applicants respectfully traverse the rejection.

As recognized by the Examiner, Tabata does not teach removing the surface of the dielectric material or the surface of the second electrode and using it as a photocatalyst material surface or photocatalyst material. However, the Examiner takes the position that one

of ordinary skill in the art would realize that a photocatalyst material is produced by Tabata's process and thus, it would have been obvious to one of ordinary skill in the art to remove the photocatalyst material from the discharge zone for use as a photocatalyst.

Applicants respectfully disagree.

Tabata discloses an **ozone generator** for generating ozone by applying a specified process to oxygen by discharge. Accordingly, an object of Tabata is to provide an ozone generator which can adequately raise an **ozone generation** efficiency. *See* [0030]. Tabata discloses that when a nitrogen dioxide gas is used as the oxide compound gas, as compared with a nitrogen gas, the **ozone generation** efficiency becomes high. *See* [0032]. Tabata further discloses that since a photocatalytic material or a material transformed into the photocatalyst is used, and the oxide compound gas is added to the raw material gas, the oxide compound gas itself has the capacity to **generate the ozone**, and the ozone can be more stably generated. *See* [0038]. Thus, the purpose of Tabata is clearly ozone generation.

A photocatalyst, such as TiO_2, is used to prevent the time-varying reduction of ozone concentration during the generation of ozone. *See* [0007]. For example, Tabata teaches that since the ozone concentration is reduced with the passage of time during the operation, the nitrogen gas is added to the raw material gas, or TiO_2 as the photocatalyst is applied to the discharge electrode surface, so that a following reaction occurs, and the time-varying reduction of the ozone concentration is prevented.

$$O_3^* + N_2 \rightarrow O_3$$

$$O_3^* + TiO_2 \rightarrow O_3$$

See [0014]. Thus, the photocatalyst is simply a component used in the process of producing

ozone.

Accordingly, in view of the purpose or objective of Tabata, which is to generate ozone, and when the entire disclosure of Tabata is considered as a whole, there is no teaching or suggestion in Tabata regarding the removal of the photocatalyst material from the apparatus of Tabata. Thus, it is respectfully submitted that one of ordinary skill in the art would not be motivated to remove the surface of the dielectric material or the surface of the second electrode and using it as a photocatalyst material surface or photocatalyst material.

Furthermore, in the present invention the oxygen gas supplied to the discharge part contains ozone. Tabata does not disclose, teach or suggest the use of an oxygen gas containing ozone is supplied to the discharge part or an apparatus including an ozonizer for supplying oxygen gas to the discharge part. In fact, the use of such an oxygen gas or ozonizer would be contrary to the object/purpose of Tabata.

For at least the above reasons, it is respectfully submitted that a prima facie case of obviousness has not been established and that claims 16 and 36 are patentable the cited art.

Additionally, claims 17-18, 22, 28, 30, 32, and 34 depend from claim 16, and thus it is respectfully submitted that these claims are patentable for at least the same reasons as claim 16.

In view of the above, withdrawal of the rejection is respectfully requested.

IV. Rejection of Claim 26 under 35 U.S.C. § 103(a)

Claim 26 is rejected under 35 U.S.C. §103(a) as allegedly being unpatentable over Tabata in view of Segawa et al. (2002/0172628).

Applicants respectfully traverse the rejection.

It is submitted that claim 26 depends from claim 19, and thus is patentable for at least the same reasons as claim 19.

Accordingly, withdrawal of the rejection is respectfully requested.

V. Rejection of Claims 19-21, 23, 29, 31, 33, 35 and 37 under 35 U.S.C. § 103(a)

Claims 19-21, 23, 29, 31, 33, 35, and 37 are rejected under 35 U.S.C. §103(a) as allegedly being unpatentable over Tabata et al. in view of Saito et al. (6,810,575).

Applicants respectfully traverse the rejection.

As recognized by the Examiner, Tabata does not teach the use of a material gas containing metal particles or a metal compound gas to be a photocatalyst element. Thus, the Examiner relies on Saito as teaching a metal compound gas to form a metal oxide in a reaction zone for use as a photocatalyst. The Examiner takes the position that it would have been obvious to use a metal compound gas in Tabata's process with an expectation of success because Saito teaches the conventionality of using a metal compound gas to form a metal oxide for use as a photocatalyst.

Applicants respectfully disagree.

Saito relates to a functional element for electric, electronic or optical device and method for manufacturing the same. Specifically, Saito discloses gasifying a metal compound having the capability to react with an oxide-forming substance to form a metal oxide. The metal compound gas is applying onto a surface of a substrate, which is placed in a reaction zone containing the oxide-forming substance and which is heated to a temperature higher than the temperature of the metal compound gas. *See* Abstract and col. 4, line 53 to col. 5, line 4. An oxide-forming substance can be, for example, water, oxygen or ammonia. *See* col. 11, lines 31-

36. More specifically, the metal compound gas reacts with an oxide-forming substance contained in the air in the reaction zone to grow the metal oxide needles on the surface of the substrate. Thus, Saito does not teach or suggest that the metal compound gas is contained in or mixed with an oxide-forming substance, such as water, oxygen or ammonia, prior to entering the reaction zone.

In contrast, in the present invention, the material gas containing metal particles or metal compound gas is contained in the oxygen gas which is supplied to the discharge zone. Since Saito fails to teach or suggest a metal compound gas is contained in oxygen, Saito does not make up for the deficiencies of Tabata.

In addition, in the present invention, the metal particles or the metal compound gas is modified into photocatalyst particles that remain in the gas and are not deposited on the substrate as in Saito.

Furthermore, it is respectfully submitted that there is no motivation to combine the references. Tabata discloses the use of oxygen having a purity of 99.99%. *See e.g.,* [0071]. Accordingly, based on the disclosure of Saito, one of ordinary skill in the art would not be lead to modify the high purity oxygen gas of Tabata with a metal compound gas, particularly since such would result in the reduction of the purity of the oxygen gas.

For at least the above reasons, it is respectfully submitted that a prima facie case of obviousness has not been established and that claims 19 and 37 are patentable the cited art.

Additionally, claims 20-21, 23, 29, 31, 33 and 35 depend from claim 19, and thus it is respectfully submitted that these claims are patentable for at least the same reasons as claim 19.

In view of the above, withdrawal of the rejection is respectfully requested.

VI. Rejection of Claim 27 under 35 U.S.C. § 103(a)

Claim 27 is rejected under 35 U.S.C. §103(a) as allegedly being unpatentable over Tabata et al. (2004/0223893) in view of Saito et al. (6,810,575) and Segawa et al. (2002/0172628).

Applicants respectfully traverse the rejection.

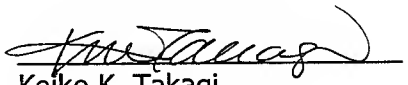
It is submitted that claim 27 depends from claim 19, and thus is patentable for at least the same reasons as claim 19.

Accordingly, withdrawal of the rejection is respectfully requested.

VII. Conclusion

In view of the above, reconsideration and allowance of claims 19-41 is respectfully requested. If any points remain in issue which the Examiner feels may be best resolved through a personal or telephone interview, the Examiner is kindly requested to contact the undersigned at the telephone number listed below. The USPTO is directed and authorized to charge all required fees, except for the Issue Fee and the Publication Fee, to Deposit Account No. 19-4880. Please also credit any overpayments to said Deposit Account.

Respectfully submitted,



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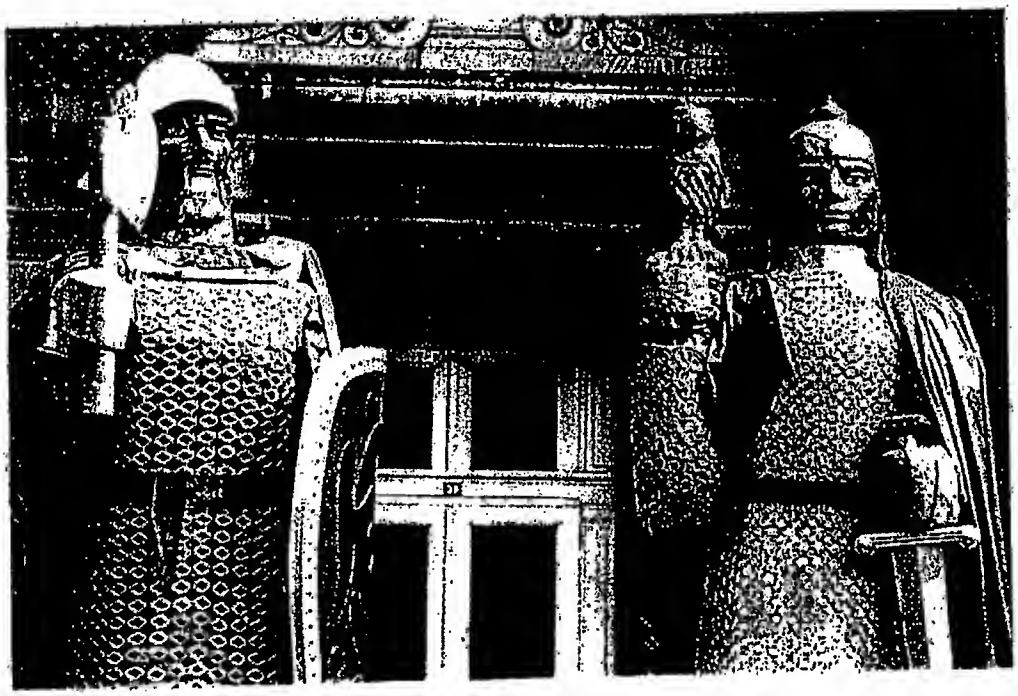

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HIGH DENSITY OZONE GENERATION IN A VERY NARROW GAP BY SILENT DISCHARGE

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Abstract

A novel structure to produce a high density ozone with a high efficiency has been developed. An extremely short gap length around 0.2mm can be formed with a high precision. In the extremely short gap length, the gas temperature in the discharge space can be limited to be sufficiently low under the high power density of the discharge. The ozone concentration as high as 200g/m^3 (in STP) is obtained with an efficiency of 98g/kWh under a high power density of 0.77W/cm^2.

Introduction

One of the key technologies to produce ozone efficiently is to keep the gas temperature sufficiently low in the discharge space. If the gas temperature rises, the production efficiency of the ozone decreases because the ozone destroying process is extremely sensitive to the temperature. The thermal diffusion equation predicts that the mean temperature rise of the gas in the discharge space between electrodes is proportional to the gap length under the condition of the same discharge power density on the electrode surface. That means when the same discharge power is inputted under shorter gap length, the gas temperature can be limited to be less. In usual high capacity ozone generating systems, a plurality of discharge tubes are used in parallel. The precision of the gap length is around to be $\pm$0.1mm. The precision of the gap length decreases, the ozone generating efficiency is to be lowered. In practical, the optimum gap length with respect to ozone generation is thought to be around between 0.8mm to 1.2mm[1],[2].

We have been developed a novel type of ozone generating apparatus in which an extremely short gap length around 0.2mm can be constructed with a high precision. In this paper, we describe the ozone yielding characteristics under narrow gap conditions, and compare the ozone yield efficiency with that of the standard gap length of 1.2mm.

Apparatus

The cross sectional view of the novel ozone generating apparatus is shown in Fig. 1. An earth electrode is made of stainless steel. A high-voltage dielectric electrode is made of alumina ceramics plate and a conduction layer which is formed by a metalized layer with a thickness of 40 micron on one side surface of the ceramics plate. A radial spacer made of metal is inserted between the earth electrode and the ceramics plate. The ceramics plate and the earth electrode form a discharge space in which the silent discharge is generated.

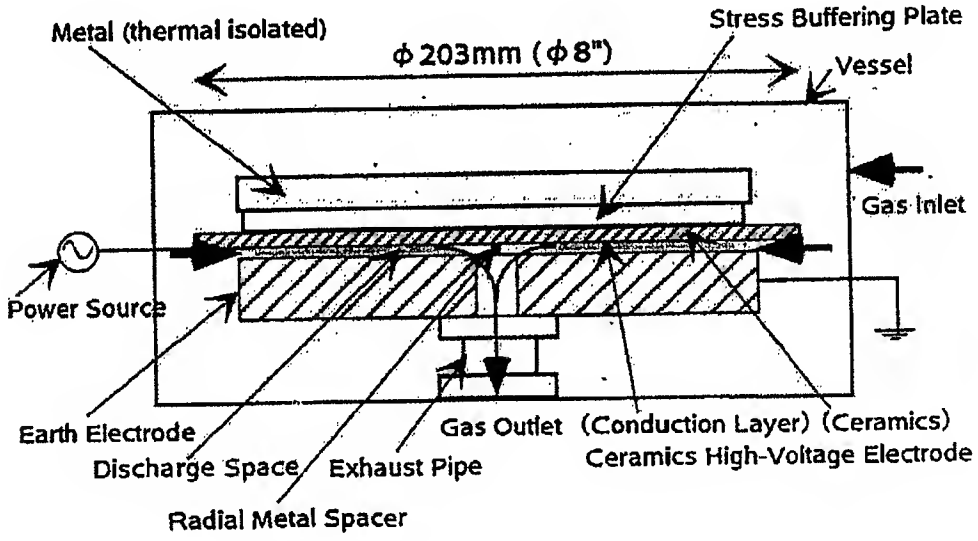

Fig. 1 Novel Ozone Generating Apparatus

The earth electrode has a hole in the center. The gas is drawn automatically into the discharge

space and evacuated through the gas exhaust pipe. Between the ceramics plate and the thermal isolated metal, a stress buffering plate is inserted. By applying a designed pressure to the thermal insulated metal, the discharge gap length is unified just to the thickness of the radial spacer . The forces generated by mechanical or thermal stress generated in the ceramics plate are absorbed by the stress buffering plate. The deterioration of the accuracy of the discharge gap length or the destruction of the ceramics plate due to a distortion of the ceramics plate can be prevented by the stress buffering plate.

Figure 2 shows the radial metal spacer and the high-voltage electrode. The radial spacer is arranged on the ceramics plate as shown in the figure. The conduction layer is formed on the reverse surface of the ceramics plate. The portions where there is no spacer are the discharge space, and the portions where there is the spacer are non-discharge parts. By this construction, the discharge space and the gas passages completely coincide to each other. Further, since the spacer occupies a large area on the whole area of the discharge space, it has become apparent that there are effects in that the accuracy of the discharge gap can be made to be uniform throughout the whole space, and the ceramics plate can be indirectly cooled through the spacer.

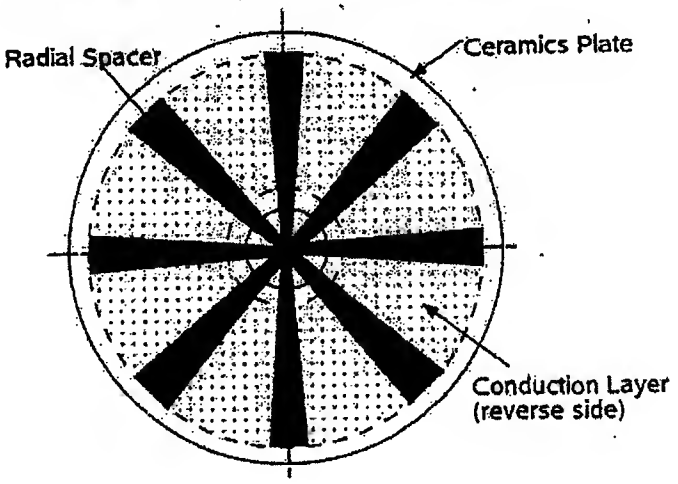

Fig. 2 Structure of High-Voltage Electrode and Spacer

Experiment

CONDITION AND METHOD

In this experiment, the area ratio of the spacer and conduction layer is set to be 20%. The effective area of discharge space is S=220cm^2. The temperature of the cooling water is 15° C. All experiments are done with oxygen gas at a gas pressure of 1.7atm. The ozone concentration is detected by means of UV absorption and checked by iodometry. The discharge power is measured by integrating the product of the discharge current I and the applied voltage V. The measured discharge power is checked by V-Q(charge) Lissajous' figure method.

RESULTS

The dependence of the discharge power W on the peak value of the applied voltage Vop to the electrode is shown in Fig.3 at various gap length of 0.2mm, 0.4mm and 0.6mm. The frequency f of the power source is 10kHz. In the figure, solid lines show the results of calculation by equation (1).

$$W = 4fC_d V^* \left\{ V_{op} - \left(1 + \frac{C_g}{C_d} \right) V^* \right\} \qquad (1)$$

where Cd and Cg are the capacitance of the dielectric electrode and that of discharge gap, and V* and Vop are the average voltage across the discharge gap and applied voltage, respectively. The values of the discharge power obtained experimentally agree well with the calculated values. A high density of the discharge power of 1.4W/cm^2 is realized at a relatively low voltage of 5kV by using a ceramics material with a high dielectric constant of 10 and a high frequency power source.

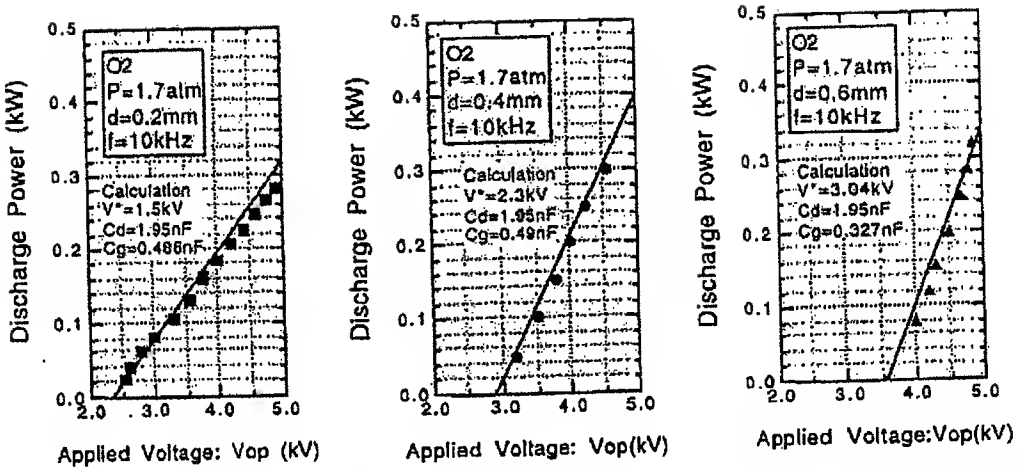

Fig. 3 Discharge power as a function of applied voltage at various gap length

Figure 4 gives the ozone concentration C_{O3} as a function of the discharge power W at various oxygen flow rates. The discharge gap is 0.2mm. The flow rate Q is expressed in STP. High ozone concentration of 200g/m^3 expressed in STP is obtained at the conditions of the oxygen flow rate of Q=1.4 litter / min and the discharge power of W=170W (discharge power density of W/S=0.77 W/cm^2). The ozone yield efficiency is about 98 gO_3 /kWh.

For comparison, one of the typical ozone yielding characteristics of commercial ozonizers with the gap length of d=1.2mm is shown in fig. 4 by a solid curve. The characteristics is measured under the following conditions; the discharge area of 2000cm^2, O_2 flow rate of 4.68l/min, gas pressure of 1.7atm, and the cooling water temperature of 15ÅãC. Initial slope for d=0.2mm is nearly equal to that for d=1.2mm at the same gas flow rate of 4.68 l/min. The saturation of ozone concentration at high power region is significantly reduced in the case of d=0.2mm in spite of a large difference in the discharge power density defined as W/S. The increase in maximum ozone concentration is thought to be caused by the improvement in cooling efficiency of the gas in the discharge space.

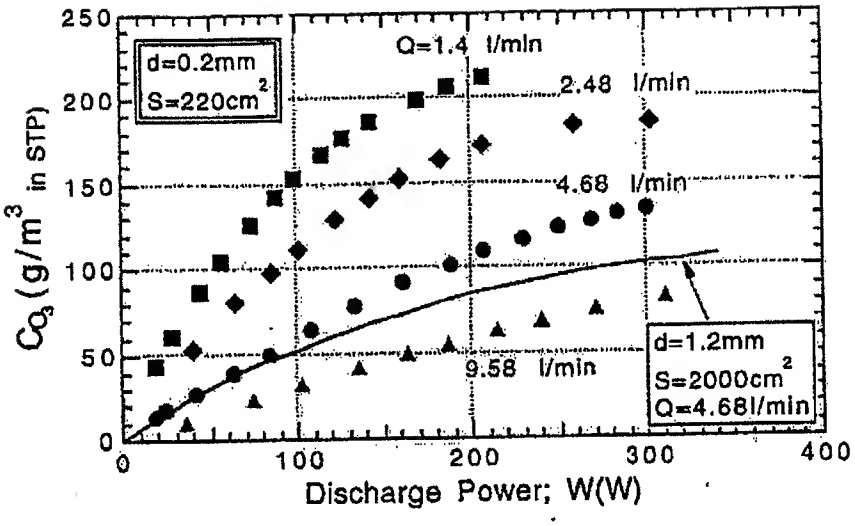

Fig.4 Ozone concentration as a function of discharge power

Figure 5 gives the ozone concentration Co3 at various gap length as a function of the normalized energy input W/Q, which is defined by the ratio of the discharge power W to gas flow rate Q (in STP) and is known to be one of key parameters on the ozone generation[3]. The maximum ozone concentration increases significantly as the gap length decreases. Roughly speaking, the increase in maximum ozone concentration is thought to be caused by the improvement in cooling efficiency of the gas in the discharge space.

In the figure, the ozone generating characteristics is very weakly dependent on the discharge power density, which is one of most important factors to determine the gas temperature. The discharge is found to be locally inhomogenized when the applied voltage is not sufficiently high against the discharge voltage. It may be necessary to redefine the discharge area S with respect to the estimation of the discharge power density.

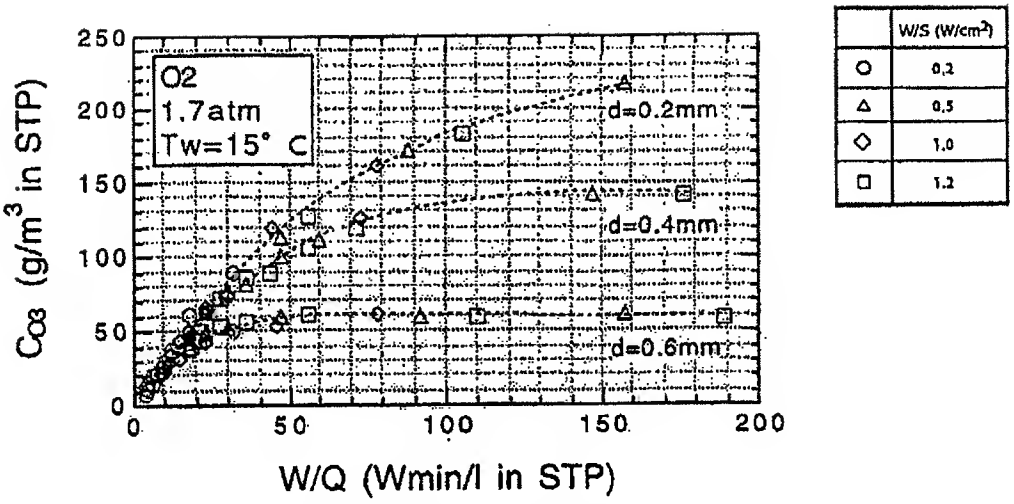

Fig.5 Ozone concentration at various gap length

Conclusion

We have been developed a novel type of ozone generating apparatus, in which an extremely short gap length around 0.2mm can be constructed with a high precision. In the apparatus, by using the stress buffering plate and the radial metal spacer, the accuracy of the discharge gap length is insured in spite of a distortion of the ceramics plate.

Realizing the ultra-short and uniform gap, the gas temperature in the discharge gap can be limited to be low, and the ozone concentration as high as 200g/m^3 is obtained with a high efficiency of 98g/kWh under a high power density of 0.77W/cm^2.

Present industrial requirements concerning the performance of the ozone generators are a high concentration and a high efficiency ozone generation and a compact design. A compact ozonizer with a large generating capacity of ozone is easily realized by stacking the modules developed here. The development of the novel ozone generator will be a significant step toward the requirements.

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PROCEEDINGS OF THE

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High Density Ozone Generation under High Electric Discharge Field

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Experimental and theoretical investigations have been carried out on the ozone yielding characteristics under the various discharge gap length from 0.05mm to 1mm. Ozone concentrations observed experimentally at various discharge power densities are explained well by a theoretical model considering the ozone dissociation by electron impact. It is concluded that the operation under a high electric field has a strong advantage to produce high density ozone because of the reduction in the population density of low energy electrons which decompose generated ozone.

1. Introduction

Ozone is an oxidizing gaseous agent, more powerful and environmentally cleaner than chlorine. Therefore, ozone is widely applied for bleaching and oxidizing purposes. But conventional ozone generators have some disadvantages such as high equipment and running cost and limited concentration of ozone. If those disadvantages are solved, ozone is expected to find much wider practical applications.

We reported high density ozone generation with high efficiency by realizing a uniform narrow discharge gap[1],[2]. In the latter paper, we pointed out that the feature of using the narrow gap lies not only in the decrease in the gas temperature rise but also in the reduction in the population densities of low energy electrons which dissociate ozone. In this paper, we report the effect of the reduction in low energy electrons on the ozone generation characteristics by using a theoretical model considering the effect of electron impact on ozone dissociation as a function of electric field strength.

2. Experiment

The conceptional structure of a new ozone generator used in this experiment[2] is shown in Fig.1. The radial spacer is inserted between the dielectric and earth electrodes. The gap length can be varied by changing the thickness of the spacer. In this experiment, the gap length is varied from 0.1mm to 0.4mm. Pure oxygen gas at pressure of 1.7atm(172kPa) is used. In order to obtain experimental results under large gaps of over 0.6mm, we also use a conventional tube type ozone generator[3].

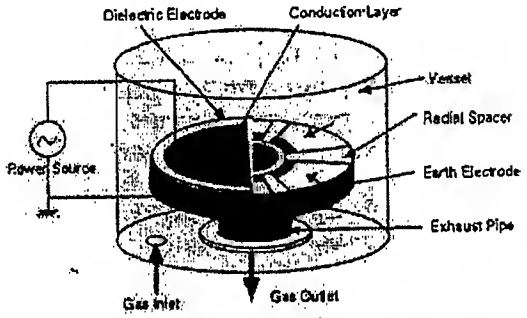

Fig.1 Configuration of the novel ozone generator

Discharge voltage V* can be evaluated from V(applied voltage) - Q(electric charge) Lissajous' figure. Figure 1 shows the experimental results of V* at 288K plotted against the nd product of gas number density (n) and discharge gap length(d). In the figure, circle symbols show the experimental results of the new ozone generator (discharge gap length d: 0.05-0.2mm, dielectric material: ceramics), while square symbols show those of the tube type ozone generator (discharge gap length d: 0.6-1.2mm, dielectric material: glass). Discharge voltages have no dependence on electric materials. The reduced electric field strength E/n (E:electric field strength) in the discharge space is obtained by dividing the discharge voltage by the nd product. It is found that the value of E/n under the operating condition of the conventional ozone generator ($nd=3.3\times10^{18}cm^2$) is around 100Td ($1Td=10^{-17}Vcm^2$) . On the other hand, by using the new ozone generator, it is possible to form discharge space with high electric

field of over 200Td.

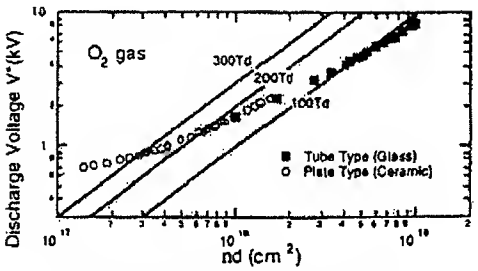

Fig.2 Reduced Electric Field Strength as a function of nd

Figure 3 shows the experimental results for several discharge gap lengths at constant gas pressure of 1.7atm. Experimental conditions such as discharge gap lengths and discharge power densities are indicated in the figure. Since gas temperature rise by discharge is proportional to discharge gap length and diseharge power density, the average gas temperatures for the four trials in Fig.3 are estimated to be constant and 312K. It is found that the maximum ozone concentration significantly increases with decreasing dischargegap length.

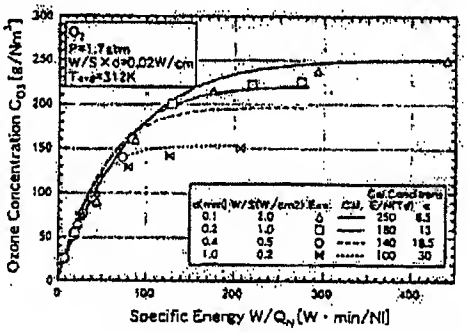

Fig. 3 Ozone generation characteristics for various discharge gap lengths at constant gas pressure and temperature

3. Discussion

The theoretical model for ozone generation used in this paper is basically the same as one report by Eliasson[4]. Except that, we consider the effect of electron impact on ozone dissociation as a function of electric field strength. Here we summarize the assumptions in this calculation: (1) the portion of energy carried by electrons is 0.55, (2) the absolute values of dissociation cross section for ozone by electron impact is 8.5 times as large as the calculated values for $O_3(^1A_2+^1A_2)$ [5]. Estimated dissociation ratio, α, expressed by the ratio k_3 to sum of k_1 and k_2 is shown in Fig.4, where k_3 is the rate coefficient of ozone dissociation and k_1 and k_2 are the rate coefficients of oxygen dissociation for the Schumann Runge system ($B^3\Sigma_u^-$) and Hertzberg system ($A_3\Sigma_u^+$), respectively.

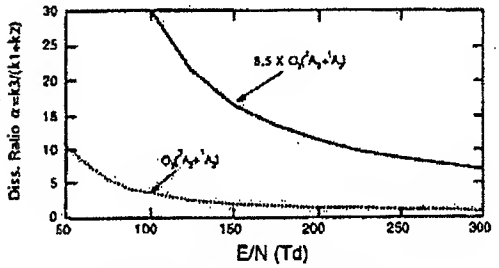

Fig.4 Estimation for Rate Coefficient of Ozone Dissociation by electron Impact

Calculated results for several discharge gap lengths are shown in Fig.3. The conditions for the calculation are also shown in the figure. The calculated results are found to be in good accordance with the experimental ones. From the comparison between the experimental and calculated results, it is concluded as follows; (1)The efficiency of ozone generation slightly decreases with decreasing gap length in the region of low ozone concentration because of the excess increase in the reduced electric field strength E/n. Operation over high E/n region of over 200Td is not so suitable to produce oxygen atoms. (2) On the other hand, the maximum ozone concentration significantly increases with decreasing discharge gap length because of the increase in E/n. On the region of high ozone concentration, the dissociation process of ozone becomes dominant. Since ozone dissociation by electron impact can be suppressed by operating under the high electric field E/n, the production of significantly high concentration ozone is possible by using the extremely short discharge gap and thus very high E/n operation.

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Theoretical and Experimental Investigation on High Density Ozone Generation by Silent Discharge

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ABSTRACT

As reported earlier, we developed a novel type of an ozone generating apparatus, in which an extremely short gap length of around 0.05mm-0.1mm can be constructed with high precision. Realizing the ultra-short and uniform gap, the ozone concentration of over 300g/Nm^3 is obtained with high efficiency of 80g/kWh under a high power density of 1W/cm^2. By using the novel ozone generator, experimental and theoretical investigations have been carried out on the ozone yielding characteristics by silent discharge (barrier discharge) under the various discharge gap length from 0.05mm to 1mm. Ozone concentrations observed experimentally at various discharge power densities are explained well by a theoretical model considering the ozone dissociation by electron impact. It is concluded that the operation under a very narrow gap length has a strong advantage to produce high density ozone because of the reduction in the population density of low energy electrons which decompose generated ozone.

1. INTRODUCTION

Ozone is an oxidizing gaseous agent, more powerful and environmentally cleaner than chlorine. Therefore, ozone is widely applied for bleaching and oxidizing purposes. But conventional ozone generators have some disadvantages such as high equipment and running cost and limited concentration of ozone. If those disadvantages are solved, ozone is expected to find much wider practical applications.

We reported high density ozone generation with high efficiency by realizing a uniform narrow discharge gap[1],[2]. In the latter paper, we pointed out that the narrow gap leads not only to the decrease in the gas temperature rise but also to the reduction in the population densities of low energy electrons which dissociate ozone. In this paper, we report the effect of the reduction in low energy electrons on the ozone generation characteristics by using a theoretical model considering the effect of electron impact on ozone dissociation as a function of electric field strength.

2. EXPERIMENT

The schematic structure of the new ozone generator used in this experiment[2] is shown in Fig.1. The radial spacer is inserted between the dielectric and earth electrodes. The silent discharge is generated through dielectric electrode in the portion where there is no spacer between both electrodes. The effective discharge area is 220cm^2. The earth electrode is cooled by water of temperature of 288K. The earth electrode has a hole in the center. Gas is drawn automatically into the discharge space and evacuated through the gas exhaust pipe.

The gap length can be varied by changing the thickness of the spacer. In this experiment, the gap length is varied from 0.05mm to 0.6mm. Pure oxygen gas at pressure of 1.7atm(172kPa) is used. In order to obtain experimental results under large gaps of over 0.6mm, we also use a conventional tube type ozone generator[3].

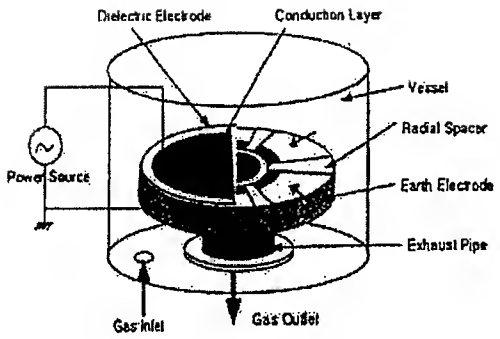

Fig.1: Configuration of the novel ozone generator

The dependence of the discharge power W on the peak value of the applied voltage Vop to the electrode is shown in Fig.2 at various gap lengths from 0.1mm to 0.6mm. The frequency f of the power source is 10kHz. In the figure, the lines show the results of calculation by equation (1).

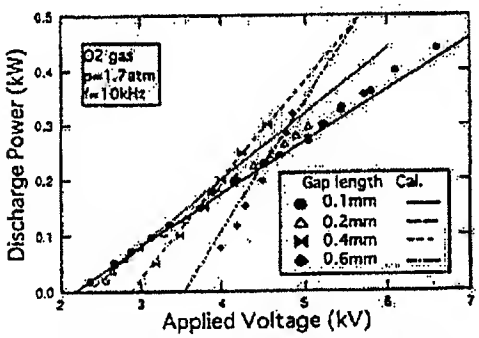

Fig.2 Discharge power as a function of applied voltage at various gap length

$$W = 4fC_d V^* \left\{ V_{op} - \left(1 + \frac{C_g}{C_d} \right) V^* \right\} \qquad (1)$$

where Cd and Cg are the capacitance of the dielectric electrode and that of discharge gap, and V* is the average voltage across the discharge gap. The values of the discharge power obtained experimentally agree well with the calculated values. A high density of the discharge power of 2.0W/cm^2 is realized at a relatively low voltage of 6.5kV by using a ceramics material with a high dielectric constant of 10 and a high frequency power source.

Discharge voltage V* can be evaluated from V(applied voltage) - Q(electric charge) Lissajous' figures. Figure 3 shows the experimental results of V* plotted against the nd product of gas number density (n) and discharge gap length(d). In the figure, the circle symbols show the experimental results of the new ozone generator (discharge gap length d: 0.05-0.2mm, dielectric material: ceramics), while the square symbols show those of the tube type ozone generator (discharge gap length d: 0.6-1.2mm, dielectric material: glass). Discharge voltages have no dependence on the materials of the electrodes. The reduced electric field strength E/n (E:electric field strength) in the discharge space is obtained by dividing the discharge voltage by the nd product. It is found that the value of E/n under the operating condition of the conventional ozone generator (nd=3.3x10^{18}cm^2) is around 100Td (1Td=10^{-17}Vcm^2). On the other hand, by using the new ozone generator, it is possible to form discharge space with high reduced electric field strength of over 200Td.

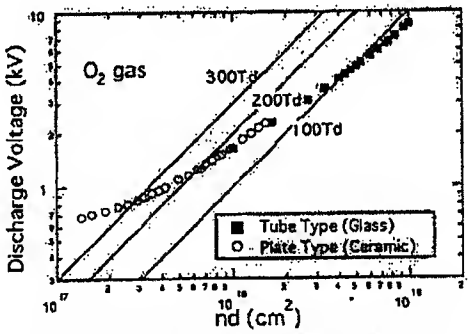

Fig.3: Reduced Electric Field Strength as a function of nd

Figure 4 shows the experimental results for several discharge gap lengths at discharge power density of 1W/cm^2. Gas pressures are optimized in each gap length. Experimental conditions such as discharge gap lengths and temperature of cooling water are indicated in the figure. It is found that the maximum ozone concentration significantly increases with decreasing

discharge gap length. The main mechanism of the increase in maximum ozone concentration lies in the improvement in cooling efficiency of the gas in the discharge space. Realizing the very narrow and uniform discharge gap of 0.05mm, the gas temperature in the discharge space can be kept sufficiently low, and ozone concentration of over 300g/Nm3 is obtained with a high efficiency of 80g/kWh under a high power density of 1W/cm^2.

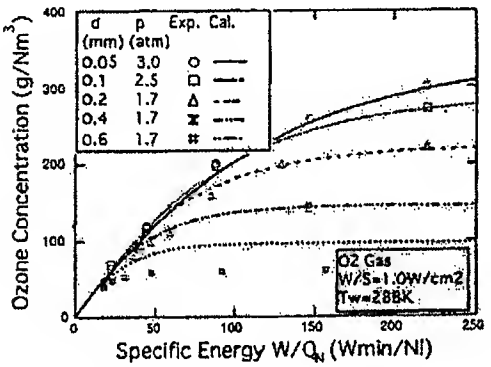

Fig. 4: Ozone generation characteristics for various discharge gap lengths at discharge power density of 1W/cm^2

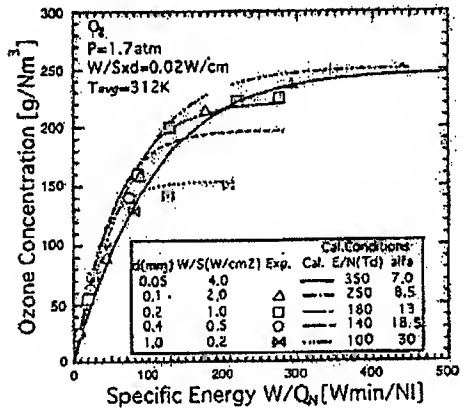

Fig. 5: Ozone generation characteristics for various discharge gap lengths at constant gas pressure and temperature

Figure 5 shows the experimental results for several discharge gap lengths at constant gas pressure of 1.7atm. Experimental conditions such as discharge gap lengths and discharge power densities are indicated in the figure. Since the gas temperature rise by discharge is proportional to discharge gap length and discharge power density, the average gas temperatures for the five trials in Fig.5 are estimated to be constant and 312K. It is found that the maximum ozone concentration significantly increases with decreasing discharge gap length. We can see clearly another advantage of using narrow gap in addition to the decrease in the gas temperature rise. This advantage of the narrow gap will be discussed in the following section.

3. DISCUSSION

The theoretical model for ozone generation used in this paper is basically the same as one report by Eliasson[4], except that we consider the effect of electron impact on ozone dissociation as a function of electric field strength. Here we summarize the assumptions in this model: (1) the portion of energy carried by electrons is 0.55, and (2) the absolute values of dissociation cross section for ozone by electron impact is 8.5 times as large as the calculated values for $O_3(^3A_2+^1A_2)$ [5].

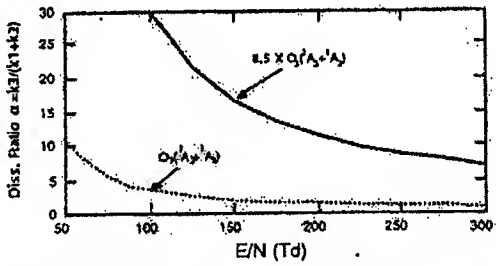

Fig.6: Estimation for Rate Coefficient of Ozone Dissociation by electron Impact

Estimated dissociation ratio, α, expressed by the ratio k_3 to sum of k_1 and k_2 is shown in Fig.6, where k_3 is the rate coefficient of ozone dissociation and $k_1$and k_2 are the rate coefficientsof oxygen dissociation for the Schumann Runge system ($B^3\Sigma_u^-$) and Hertzbergsystem ($A_3\Sigma_u^+$), respectively[6].

Calculated results for several discharge gap lengths are shown in line form in Fig.4 and Fig.5. The conditions for the calculation are also indicated in the Fig.5. The calculated results are found to be in good accordance with the experimental ones. From the comparison between the experimental and calculated results, it is concluded as follows; (1) The efficiencyof ozone generation slightly decreases with decreasing gap length in the region of low ozone concentration because of the excess increase in the reduced electric field strength E/n. This slight decrease is clearly confirmed in the case of the gap length of 0.05mm. Operation over high E/n region of over 200Td is not so suitable to produce oxygen atoms and to produce low concentration ozone. (2) On the other hand, the maximum ozone concentration significantly increases with decreasing discharge gap length because of the increase in E/n. Over the region of high concentration ozone, the dissociation process of ozone becomes dominant. Since ozone dissociation by electron impact can be suppressed by operating under the high electric field E/n, the production of significantly high concentration ozone is possible by using the extremely short discharge gap and thus very high E/n operation.

4. CONCLUSIONS

We developed a novel type of ozone generating apparatus, in which an extremely short gap length of around 0.05mm-0.1mm can be constructed with high precision as reported earlier. Realizing the ultra-short and uniform gap, the ozone concentration of over 300g/Nm^3 is obtained with a high efficiency of 80g/kWh under a high power density of 1W/cm^2.

By using the novel ozone generator, experimental and theoretical investigations have been carried out on the ozone yielding characteristics by silent discharge (barrier discharge) under the various discharge gap length from 0.05mm to 1mm. It is found that maximum ozone concentration significantly increases with decreasing discharge gap length under the condition of identical gas temperature. Ozone concentrations observed experimentally at various discharge power densities are explained well by the theoretical model considering the ozone dissociation by electron impact. It is concluded that the operation under a very narrow gap length has a potential advantage to produce high density ozone because of the reduction in the population density of low energy electrons which decompose generated ozone and as well as because of the decrease in the gas temperature rise.

5. ACKNOWLEDGMENTS

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